MATHEMATICAL MODELING OF TURBULENT REACTING PLUMES

Executive Summary

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"Continued Development

of a Mathematical Modeling Capability

in Photochemical Air Pollution:

Reacting Plumes"

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EXECUTIVE SUMMARY

In the A.R.B. Report titled "Mathematical Modeling of Turbulent Reacting Plumes," a new, comprehensive model (TRPM) for chemically reacting plumes, is presented, that accounts for the effects of incomplete turbulent macro- and micro- mixing on chemical reactions between plume and ambient constituents. Comparison with experimental data shows the TRPM capable of quantitatively predicting the retardation that is imposed on the evolution of fast nonlinear plume chemistry by the atmosphering mixing processes.

The downwind evolution of atmospheric plumes involves a variety of physical and chemical processes, single phase and multi-phase, linear and non-linear, interacting or non-interacting, that are characterized by a variety of time scales and therefore are relevantnt during different, shorter or longer, periods of the dispersion (Figure 1). Whereas simple existing models have been shown to describe more or less adequately linear (or quasi-linear) phenomena, there are significant questions regarding the proper description of nonlinear processes (commonly nonlinear chemical rates). This is especially true for the first few kilometers of downwind dispersion, where these processes are strongly affected by the state of incomplete and nonideal mixing between emissions and the ambient. Most existing reacting plume models are based on very simplistic descriptions of atmospheric mixing (Figure 2). Thus, emissions from large industrial stacks are typically assumed to mix completely and instantaneously with a relatively large volume of ambient air as soon as they leave the source. Then chemistry is assumed to take place in a homogeneous and deterministic manner inside a computational volume where emissions and ambient species are assumed thoroughly mixed down to the molecular level. This significant *artificial dilution* of

the emissions, and also the common practice of ignoring (or simplifying the structure of) gradients of expected concentration profiles inside the plume (i.e. the state of "plume macromixing"), as well as the existence of local, instantaneous, concentration fluctuations about these expected profiles (the magnitude of which determines the state of "plume micromixing"), may lead to significant deviations between calculated and actually observed mean concentration values. Furthermore, the use of dispersion schemes that are (explicitly or implicitly) relevant to a time-averaged picture of the dispersion process (e.g. the common Atmospheric Diffusion Equation or K-theory), is in principle inconsistent with the simultaneous description of fast nonlinear chemical kinetics (typical of the NO_x - O_3 system), and may also lead to significant errors when applied beyond the limits defined by certain scales that characterize the overall reaction and diffusion system.

The above problems have been considered in detail in this report and a new, comprehensive "Turbulent Reacting Plume Model" (TRPM) has been developed in order to address them. This model takes into account the effects of both mean gradients and fluctuations on the evolution of plume chemistry, for those ranges of the dispersion where they have a significant effect. To ensure applicability, the TRPM was constructed so as to be relatively simple from a computational point of view, adhering to a first order closure scheme for the description of the turbulent atmospheric diffusion process, the necessary higher order closure being confined to the formulation of the turbulent kinetic terms.

Instead of simply adopting already available (but not sufficiently developed or computationally simple) theories and techniques for describing the phenomena relevant to turbulent reacting plumes, development and implementation of the TRPM was based on the introduction of new, original, concepts and methods relevant to distributed parameter stochastic reacting systems. Thus the "phenomenal extent of reaction" (in connection with fictitious inert surrogates of the emission species) is introduced to reduce much of the computational burden involved in the reaction-dispersion calculations, the "concentration field splitting technique" (based on an

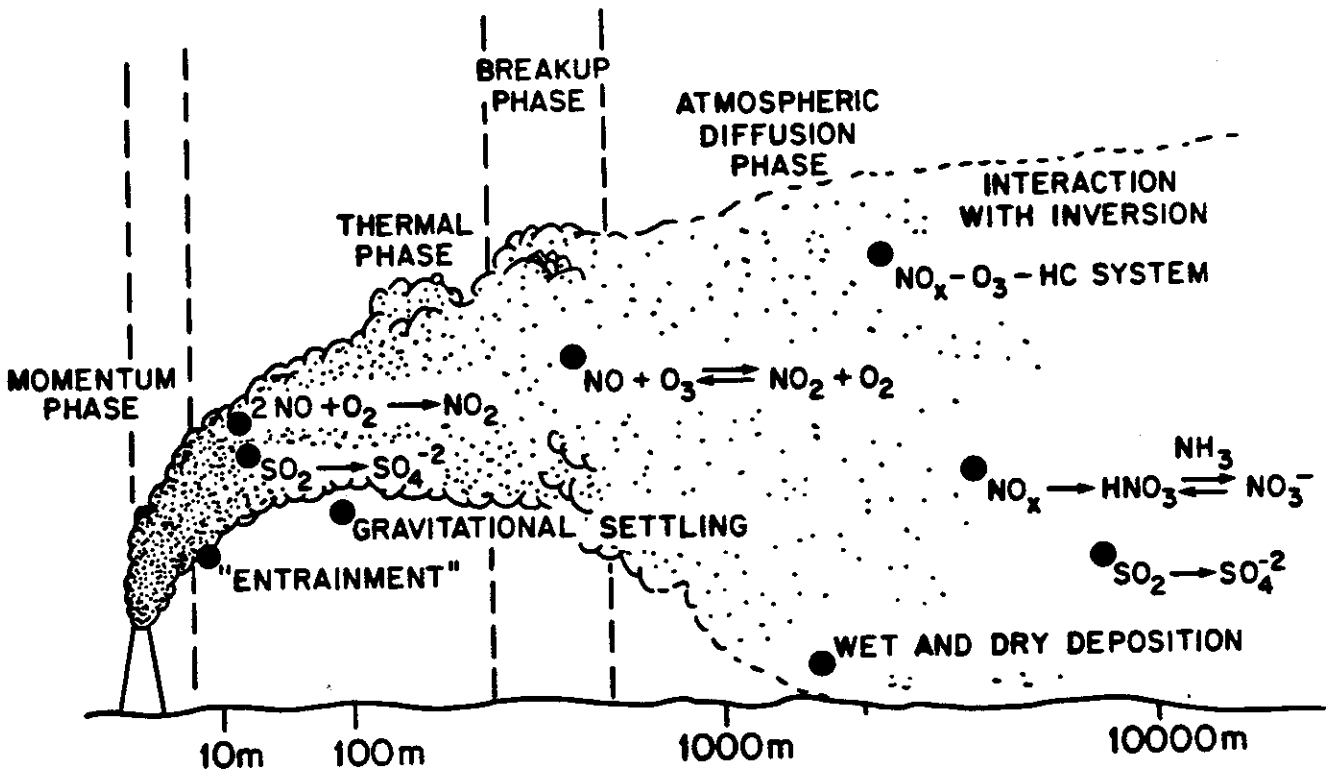

Figure 1

Schematic Representation of the Evolution
of the Various Typical "Phases" of Plume Physics and Chemistry
(and of the spatial scales associated with these phases).

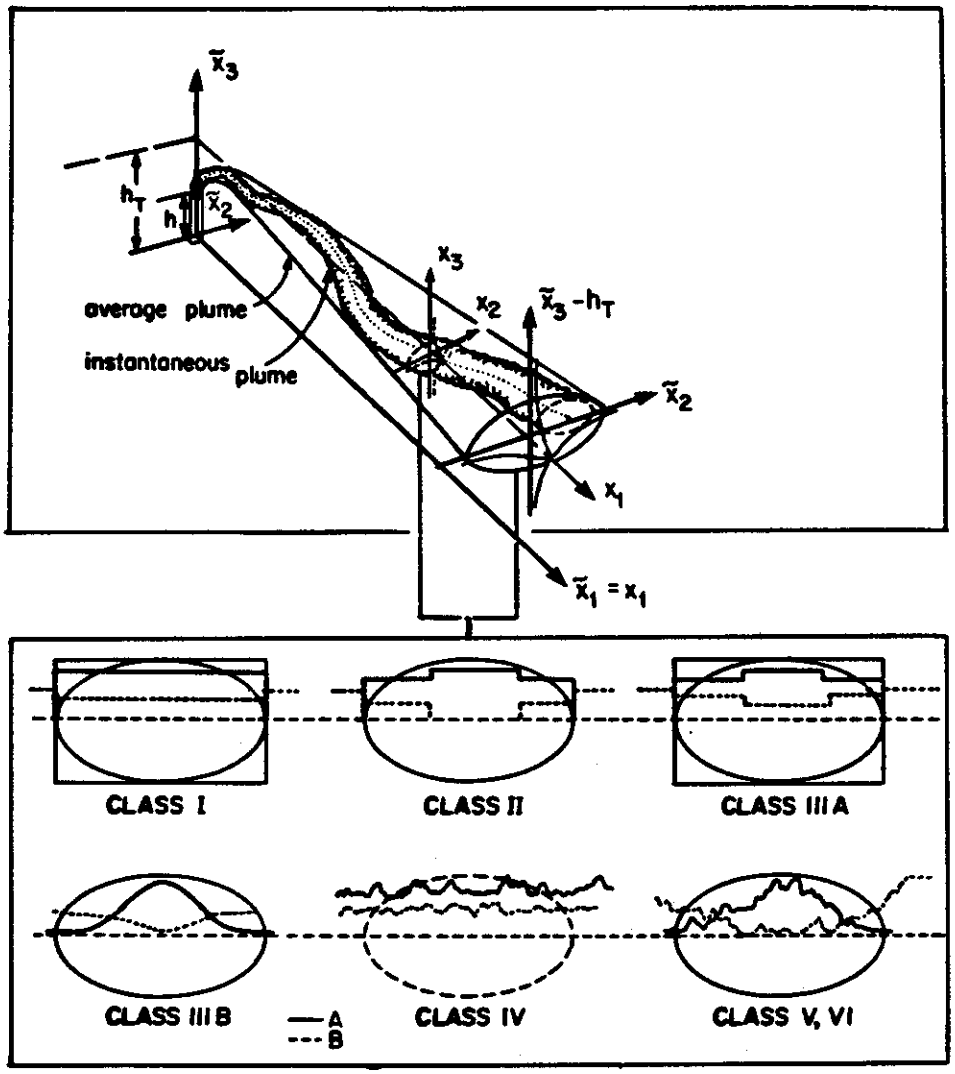

Figure 2

Schematic Representation
of the Basic Characteristics of Existing Reactive Plume Models
(according to the classification of Section 3)

The expanding plume is identified as a special control volume inside the atmospheric boundary layer (with the exception of the models of Class IV that consider uniform values of mean concentrations over the entire boundary layer), corresponding to either instantaneous plume realisations or (more often) to the time averaged envelope. (For models relevant to instantaneous realisations the mean concentrations must be ensemble averages and they can be in steady state only with respect to a frame of reference that follows the meandering centerline.) The cross-wind section of the plume is usually assumed to have elliptical shape (or rectangular in some of the models of Class I and IIIA); concentration profiles of the emitted and the ambient species (A and B respectively) that are typical in the various models are shown: Classes I to IIIB ignore the state of micromixing (local fluctuations) and assume profiles that are uniform (Class I), sectionally uniform (Class II, IIIA) or Gaussian (Class IIIB). Class IV takes into account fine scale fluctuations but ignores variations in the mean profiles. Finally Class V (and VI) consider the effects of both macromixing and micromixing.

integral functional representation of the random concentration fields) is introduced to allow chemical closure - for an arbitrarily fast reaction - at the same order as of the kinetics, the "localized production of fluctuations method" is introduced (as a means for describing the evolution of the variance of plume concentration) to obtain closed analytical solutions for the plume segregation and closed expressions for second order turbulent kinetics, etc.

Furthermore, the TRPM is modular in nature, consisting of independent components for the description of non-interacting processes (i.e. bulk plume motions and "internal" mixing and reaction), and allowing for the use of different alternatives on the level of approximation of the phenomena involved (Figure 3).

The core of the TRPM (the "master module") consists of the evolution equations for reaction progress variables appropriate for evolving, spatially varying systems (i.e. the local phenomenal extent of reaction). These equations estimate the interaction of mixing and chemical reaction and require input parameters characterizing internal plume behavior, such as relative dispersion and fine scale plume segregation. Expected concentrations etc. are estimated in a frame following the expected motion of the instantaneous plume. The calculations of the master module are extended by the components of the "complementary module," that account for the effects of bulk plume motions (buoyancy induced rise, random meandering), and thus allow the estimation of fixed frame expected concentrations and time averages. Furthermore, "peripheral modules" supply various input parameters required by the master and complementary modules.

The main body of the present report is organized into two parts:

Part IA (Chapters 1 to 3) contains:

- A general discussion and a literature survey of reactive plume models,
- a detailed description of the TRPM structure, and
- comparisons of calculations with measurements.

Calculations performed with the TRPM are compared with the experimental data of Builtjes (1981) for the reaction between NO in a point source plume and

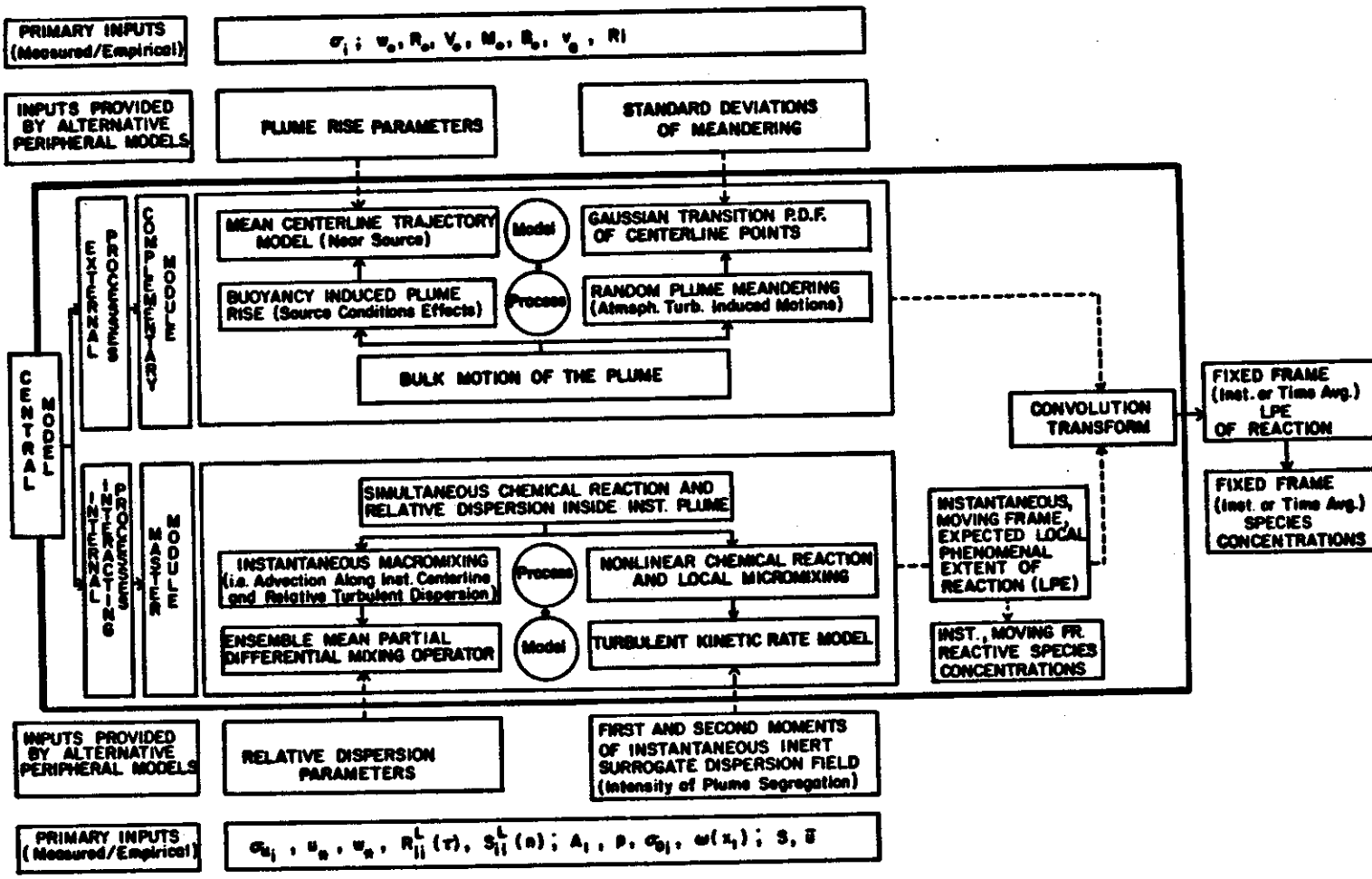

Figure 3

Structure of the TRPM

ambient O_3, taking place in a wind tunnel simulating a neutral atmospheric boundary layer. The comparison shows the TRPM capable of quantitatively predicting the retardation imposed on the evolution of nonlinear plume chemistry by incomplete and non-ideal mixing (see, e.g., Figures 4 and 5).

Part IB (Chapters 4 to 7) contains systematic studies on the turbulent dispersion and reaction phenomena and plume dynamics, thus exposing in detail the underlying concepts and methods relevant to turbulent reactive plume modeling. The fundamentals of various new formulations that were developed to describe in-plume phenomena are included here.

A somewhat more specific outline of the contents of Part IB follows:

Chapter 4 contains

- an introduction to the problem of modeling nonlinear chemical reactions in turbulent flows, including a general literature survey and classification of existing methods,
- an exposition of the fundamentals of the Eulerian Statistical Approach for both direct and indirect chemical closure methods,
- a detailed presentation of the "concentration field splitting method," that is the particular indirect closure technique employed in the formulation of the TRPM.

Chapter 5 contains

- an introduction to the problem of modeling concentration fluctuations in point source plumes, including a brief literature survey,
- an exposition of the fundamental concepts and problems of a meandering frame Eulerian Approach for modeling the instantaneous "internal" plume concentration variance, with extensive discussion of the self similarity concept,
- a detailed discussion of the new "Localized Production of Fluctuations Method," that is the first choice for use with the TRPM.

Chapter 6 contains

- an introduction to the formal description of fluid particle dispersion (in both

Comments on Figures 4 and 5

Both calculations and data are presented in dimensionless form, as ratios of concentrations, and therefore give directly the dependence of conversions on downwind distance or travel time. The farthest downstream available measurements in the wind tunnel were taken at 5 m, which at a wind speed of 0.4 m/s corresponds to a travel time of 12.5 s. The respective quantities for a field situation would be approximately a 2.5 km downwind distance (or 7.5 min of travel time) with an average wind speed of 5.7 m/s.

Figure 4 contains the measured values of the ratio of NO to total NO_x as well as three calculated curves:

(a) *turbulent kinetics calculations,* of $\langle c_{NO} \rangle / \langle c_{NO_x} \rangle$ i.e. results from the TRPM considering the effects of both macromixing and micromixing, with parameters estimated as described in the previous section,

(b) *conventional kinetics calculations,* i.e. results from the common reaction–diffusion equation (to which the TRPM reduces for $I_s = 0$), that incorporate solely the effects of macromixing, and,

(c) *modified Gaussian plume calculations,* i.e. results from the pseudo-linear reaction assumption (equation (3.3-10) of the report) which views plume macromixing in a very simplistic manner, that is with ozone profiles unaffected by the plume, and, of course, ignores micromixing effects. The same dispersion parameters were used in all the three different models. The 50% conversion time predicted by the TRPM is 9.0 s (of the order of 5.3 min for the comparable field situation), in very good agreement with the wind tunnel measurements. The respective calculations from the conventional reaction–diffusion equation gave a 50% conversion time equal to 6.5 s (3.8 min in the field stuation), i.e. about 28% shorter than the observed time, and the modified Gaussian plume model predicted 4.75 s (2.8 min in the field), that is almost 47% less than the observed value.

Figure 5 shows the measurements of the ratio of O_3 concentration (under reactive conditions) to the corresponding background value along the centerline, again compared with calculations that assume turbulent and conventional kinetics. (The relevant assumption of the modified Gaussian plume model is that $\langle c_{O_3} \rangle = \langle c_{O_3} \rangle^{\text{env}}$ and the ratio shown in Figure 5 is always unity).

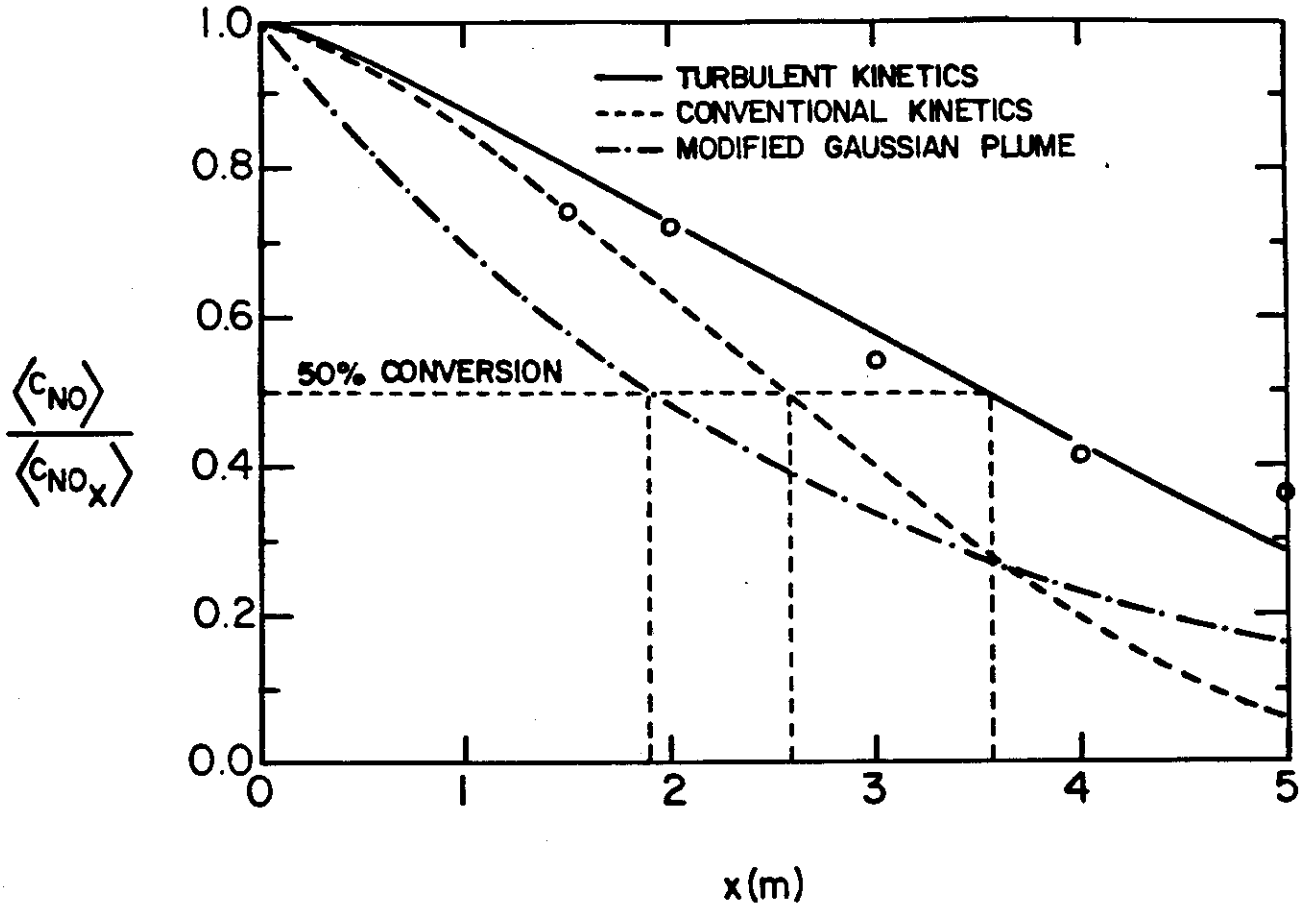

Figure 4

Ratio of NO to Total NO_x Concentration along the Plume Centerline:
Data of Builtjes (1981) and Calculations
(i) for Turbulent Kinetics (the complete TRPM equations),
(ii) for Conventional Kinetics (ignoring the effects of micromixing), and
(iii) from a Modified Gaussian Plume Model.

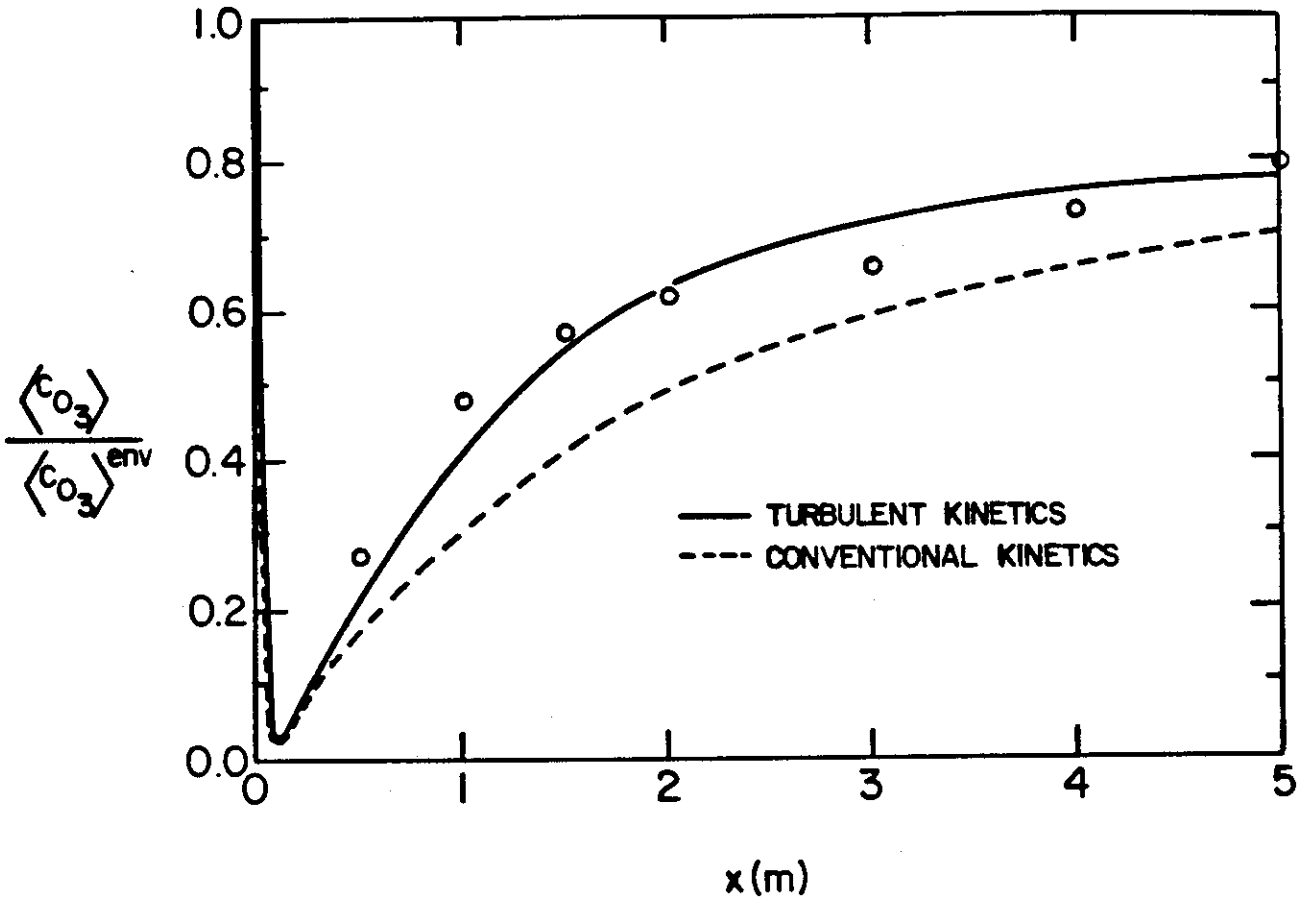

Figure 5

Ratio of O_3 Concentration (under reactive conditions)
to the Corresponding Background Value ($\langle c_{O_3} \rangle^{env}$) along the Plume Centerline:
Data of Builtjes (1981) and Calculations
(i) for Turbulent Kinetics (the complete TRPM equations), and
(ii) for Conventional Kinetics (ignoring the effects of micromixing).

inertial and non-inertial frames) via stochastic and deterministic Green's functions,

- a discussion of the modified A.D.E. in relation to other dispersion models,
- a review of time-domain methods for determining relative dispersion parameters (including similarity analysis, Langevin equation methods and conditioned motion methods),
- a discussion of spectral methods for determining relative dispersion parameters and presentation of an iterative filtering algorithm that utilizes observed atmospheric spectra for this objective.

Chapter 7 contains

- a discussion of the initial phases of plume dispersion, and the scales and properties that are relevant to each phase,
- a brief overview of methods employed to model plume rise,
- a presentation of the models of Briggs and Schatzmann that are suggested as the "simple" and "comprehensive" alternatives, respectively, for use with the TRPM.

References

Builtjes, P.J.H. (1981) "Chemically Reacting Plume Experiments for the Wind Tunnel," Netherlands Organization for Applied Scientific Research, Division of Technology for Society, Ref. No. 81-013563, File No. 8710-20330.